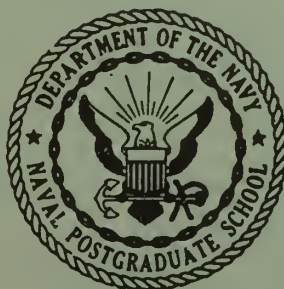


ORIENTATED SINGLE CRYSTAL
COPPER FIBERS BY
A STRAIN - ANNEAL TECHNIQUE

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AND
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UNITED STATES NAVAL POSTGRADUATE SCHOOL



THESIS

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Submitted in partial fulfillment of
the requirements for the degree of

MASTER OF SCIENCE
IN
PHYSICS

United States Naval Postgraduate School
Monterey, California

1 9 5 7

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ABSTRACT

A vacuum furnace for high temperature anneal of metal fibers with a moving gradient was designed and constructed. The design specifications are detailed. Systematic evaluation of the variables governing the longitudinal crystal growth of individual copper crystals in a 16 mil fiber was carried out. Single crystals approximately 40 mils in length were obtained by a 1010° C. anneal, specimen velocity of 0.005 mm./second through a 10 cm. furnace, with 2.5 grams of tension on specimen. Specimen had undergone 92% diameter reduction by cold-drawing prior to anneal.

PREFACE

The investigation of crystalline lattice imperfections and their effect on the physical properties of material has been the subject of much post World War II research in the field of solid state physics. Modern experimental tools, such as x-ray diffraction, the electron microscope, particle accelerators, etc, permit experimental investigation into the mechanism of lattice imperfections that could not previously be examined. The effects of these imperfections on the gross physical properties of the bulk material can be studied and evidence gathered concerning their interrelationships. This evidence is leading to a fuller understanding of the mechanisms involved although they are not yet completely determined.

Studies have been in progress at the United States Naval Postgraduate School since 1953 with the objective of learning more about the basic mechanisms involved in damage to crystal lattices, how such damage may repair itself and what effect these imperfections have upon the physical and mechanical properties of the material. This work has been done in three major fields;

- a. Plastic deformation of crystals
- b. Vapor deposition of thin metal films
- c. Electron damage to crystal lattice.

Plastic deformation studies involve the introduction of imperfections by cold-working the metal under study and observing the effects on physical properties. These imperfections are usually inserted under low temperatures so that diffusion of the defects is not a consideration. Pulse anneals may then be used to investigate the recovery that occurs as thermal diffusion allows the imperfection to migrate through the crystal.

Vapor deposition of thin metal films is a technique used to learn more about the environment required for the growth of a metal crystal. Single nucleating centers are formed by evaporation of metal onto a suitable surface and the mechanism of crystal growth about these centers is observed.

Electron damage to a crystal lattice is a very useful tool for the study of imperfections. The energies involved result in the production of single interstitial-vacancy pairs without the added complication of thermal spikes arising from higher energy bombardment. These interstitial-vacancy pairs appear to pin dislocations inserted by plastic deformation. The modification of physical properties by this pinning forms the basis of some experimental work.¹

Experiments in this field performed by Powell and Sellers on the effect of electron damage on the shear modulus of copper utilized the horizontally mounted, two million electron volt Van de Graaff accelerator located at the Naval Postgrad-

¹J. W. Glenn, Advances in Physics (a quarterly supplement of the Philosophical Magazine) Vol. 4., Number 16, pp 381-478, October 1955.

uate School. They continued experimentation performed by Dieckamp and Crittenden¹ with 20 million electron volt deuterons, and by Dieckamp² with one million volt electrons. Powell and Sellers irradiated pure copper specimens at liquid nitrogen temperatures and noted a large increase in the shear modulus.³ Their work was supervised by Dr. Crittenden, now of the faculty of the Postgraduate School.

Following completion of this experiment, it was decided to utilize the remaining few copper specimens (99.999% pure, singly annealed for $[322]$ crystal orientation and large grain size) for an investigation of irradiation effects in intentionally cold-worked specimens. Sellers performed two such experiments in June of 1956 with the assistance of the authors. Results of these runs were included in a presentation to the December 1956 meeting of the American Physical Society.⁴

Further experimentation along these lines would require more specimens and their fabrication is the subject of this paper. It was decided to grow single crystal specimens with

¹H. Dieckamp and E. C. Crittenden, Jr, Shear Modulus of Irradiated Copper, Phys. Rev. 94, 1418 (1954)

²H. Dieckamp, Shear Modulus Recovery of Electron Irradiated Copper, Phys. Rev. 98, 1531 (1955)

³D. A. Powell, Jr. and H. S. Sellers, The Effects of Electron Damage on the Shear Modulus of Copper, Thesis Project, U.S.N.P.G.S., 1956

⁴D. A. Powell, H. S. Sellers, E. A. Milne and E. C. Crittenden, Jr, Equipment for Observing Shear Modulus of Copper under Irradiation, Bulletin of the American Physical Society, Vol. 1, page 379, 27 December 1956.

[111] crystal orientation. The apparatus described herein was constructed and tested at the United States Naval Postgraduate School during the period June 1956 through April 1957 in order to produce these specimens.

The authors wish to express their appreciation for the assistance and encouragement given them by Professor E. C. Crittenden, Jr, to whom credit for the initial concept of the apparatus should be given and to Professor J. R. Clark for his help with the x-ray diffraction testing. They also wish to acknowledge the valuable technical assistance given them by Mr. M. K. Andrews, Mr. D. Clark and Mr. K. C. Smith.

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CHAPTER I

INTRODUCTION

1. Specimen requirements

Previous investigation in the field of irradiation effects on the torsional shear modulus of copper performed at the United States Naval Postgraduate School utilized copper specimens ten mils in diameter. These specimens were singly annealed and exhibited an average grain size less than the fiber diameter with a $[322]$ fiber orientation. This grain size resulted in many crystal boundaries in the section of the specimen which was irradiated. The effect of these boundaries is not fully understood and their presence complicates the interpretation of experimental results. The $[322]$ fiber orientation is 11 degrees away from the 111 orientation desired for this study. The $[111]$ orientation is preferable as the orientation function is constant for this plane and the torsional shear modulus is isotropic. Previous specimens also had an oxide coating which had an undetermined effect of the torsional shear modulus.

It was decided that specimens produced for future use should, ideally, possess the following properties:

- a. absolute purity
- b. surface free from oxidation
- c. $[111]$ fiber orientation
- d. single crystal structure.

The requirement for absolute purity was approached by procuring spectrographically standardized copper rods from Johnson, Matthey & Co., Limited, of 73/83 Hatton Garden, London. The report furnished with these 5 mm. diameter rods shows impurities as follows:

- a. Two parts per million of Ni, Fe
- b. One part per million of Ag
- c. Less than one part per million of Mn, Ca, Mg, Si, Na
- d. No trace of Al, As, Au, B, Ba, Be, Bi, Cd, Co, Cr, Ga, Ge, Hg, In, Ir, K, Li, Mo, Nb, O, Os, P, Pb, Pd, Pt, Rb, Re, Rh, Ru, Sb, Se, Sn, Sr, Ta, Te, Ti, Tl, V, W, Zn, Zr.

Oxidation of the specimen may be reduced if all annealing is done in hard vacuum or in an inert atmosphere.

When polycrystalline specimens in which the grains are randomly oriented undergo deformation, the orientation of the grains is altered. This alteration frequently consists of a reorientation in which the random character is changed to some preferred orientation. The deformation also stores elastic energy in the grains of the metal which will appear upon annealing as the energy required to promote growth of new grains. Bassi¹ found a single $[111]$ texture in copper wire after a 950° centigrade anneal following a 90-97 per cent deformation and concluded that this texture results from high deformations and annealing at high temperature. The specimens used were

¹Bassi, G., Recrystallization Textures in Copper Wire, Journal of Metals, pp 753-4, July 1952.

99.90 per cent pure copper with deformation inserted by cold-drawing from 16 mm. to 1 mm. diameter.

Hibbard¹ found both $[100]$ and $[111]$ orientation after an 86 per cent diameter reduction but found primarily a $[111]$ orientation after a 96.4 per cent reduction. He concluded his discussion by saying

In summary, the deformation texture of cold-drawn copper wire still may be described as primarily a single $[111]$ texture after a sufficient amount of drawing, such as 96.4 per cent reduction in diameter.

In the preparation of the copper specimens described herein, deformation resulted from cold-drawing the 5 mm. copper rods, previously described, to a diameter of 0.016 inches by 60 draws through a plate wire die. The 16 mil diameter represents the smallest die which was available and is a 92 per cent reduction in diameter.

Growth of new grains during anneal occurs as a result of the release of elastic energy stored prior to the annealing process. A thermal gradient will allow all stored energy to be released in a short section of the fiber and will favor the continued growth of one crystal over the simultaneous growth of several. Hence, a sharp thermal gradient will implement the growth of a single crystal.²

¹Hibbard, W. R., Deformation Texture of Cold-drawn Copper Wire, Journal of Metals, page 467, June 1951

²Private communication between Professor E. C. Crittenden, Jr., U.S.N.P.G.S. and Dr. Lowe, General Electric Corporation.

2. Apparatus requirements

The apparatus to be designed for the production of these copper specimens must fulfill certain requirements if it is to perform satisfactorily. These requirements include:

- a. capability for a high temperature anneal in hard vacuum or in an inert atmosphere for varying lengths of time,
- b. some means of achieving relative motion between the specimen and the furnace,
- c. a means of producing a sharp thermal gradient at the ends of the furnace,
- d. a means of accurate temperature control, including remote indication and recording.

Serious consideration was given to the use of a mobile furnace and a stationary specimen. Several models were made but the difficulty of supplying large electric currents to a mobile furnace using uninsulated wires in a hard vacuum, coupled with the difficulty of achieving a thermal gradient under these conditions, forced their rejection. Therefore, a stationary furnace and a mobile specimen were incorporated in the design and proved very satisfactory although this limits the specimens to half the length possible with a mobile furnace.

The thermal gradient and temperature recording criteria were met and are discussed in detail in Chapter II.

3. Design considerations

The system variables which could possibly have an effect on crystal growth or orientation include:

- a. temperature of the specimen
- b. pressure in the system
- c. composition of the gaseous environment
- d. duration of anneal
- e. tension on the specimen
- f. sharpness of the thermal gradient.

All of the above variables were considered in the design of the equipment described in Chapter II, and a measure of control of each was afforded. Another consideration, of less immediate importance but which nevertheless had an effect on the equipment was that, ideally, the equipment should possess the maximum possible flexibility. This would make it easy to adapt the apparatus to other allied procedures in the field of high temperature, vacuum annealing.

CHAPTER II

EXPERIMENTAL APPARATUS

1. General Description of Equipment

Taking the requirements for specimen treatment into consideration, the design of the experimental equipment evolved into the general form shown in Figs. 1 and 2. As it is shown in Fig. 3, the system is built around a framework. This framework was obtained commercially. To provide a surface which would give structural rigidity and sufficient space from which the various portions of the system could be constructed, a mounting table of half inch aluminum plate was attached to the top of the framework.

Fig. 3 shows the manner in which the pumping portion of the vacuum system is attached to the under side of the mounting table. Immediately above the mounting table is the cold trap for the vacuum system and the drive portion of the mechanical system. Above this is a one inch aluminum base plate which serves as a base for the working section. These details can be seen most clearly in Fig. 2. The working section is enclosed by a six inch inside diameter pyrex tube. The end of the main tube is capped off by the end plate and compression collar shown in Fig. 4.

2. Vacuum System

The entire pumping section of the equipment is located beneath the mounting table. Fig. 3 shows the details of this

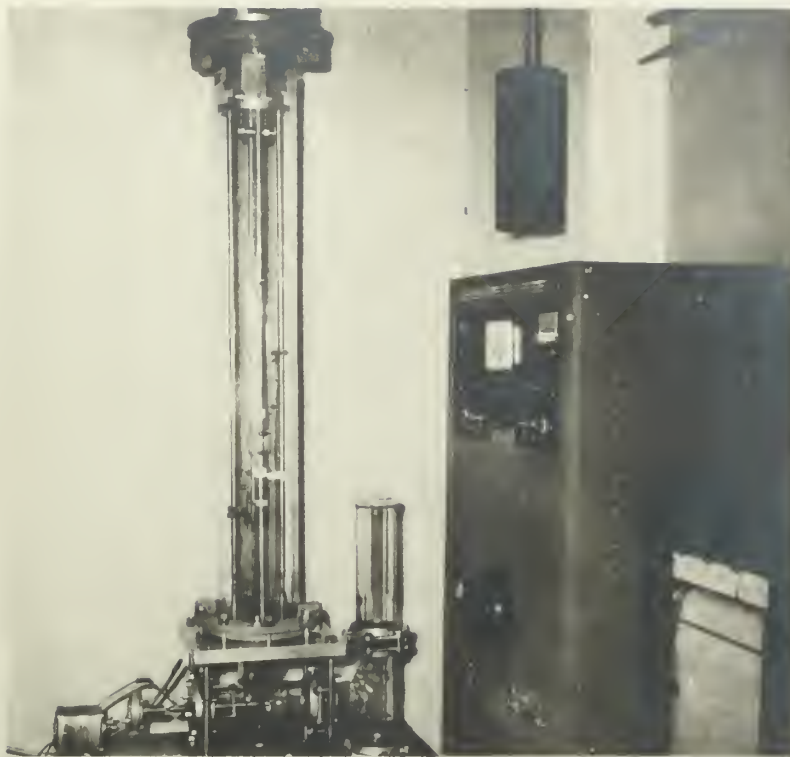


Figure 1
Side View Of Equipment

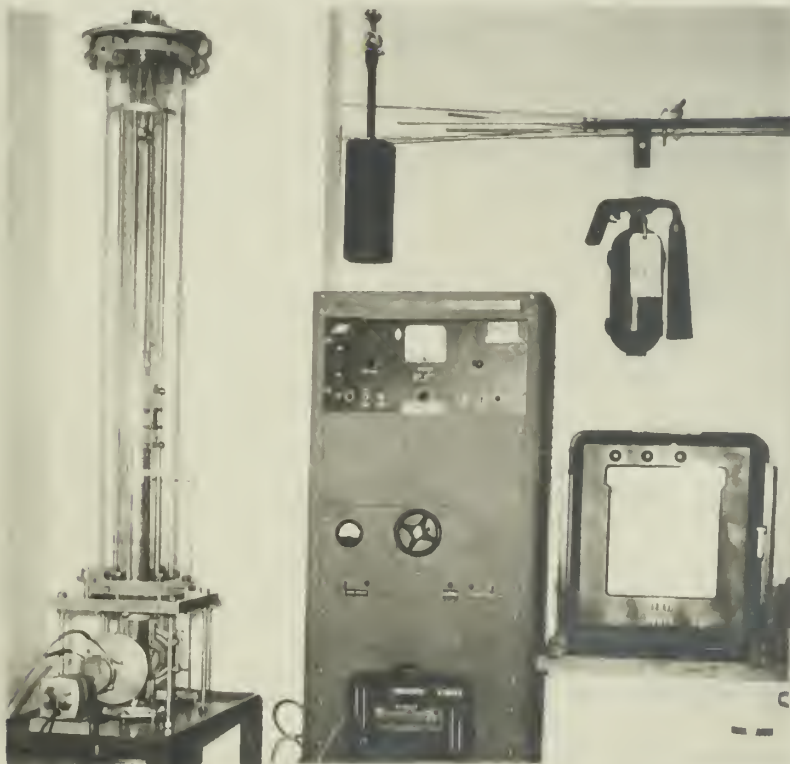


Figure 2
Front View Of Equipment

section. The fore-pump is coupled into a four inch water cooled diffusion pump. The diffusion pump is connected to a four inch cold water baffle. The baffle was inserted here to reduce the amount of oil vapor which could pass from the diffusion pump up into the working section of the equipment. The cold water baffle is secured to the under side of the mounting table. Next, in the vacuum system, is the cold trap which can be seen in Fig. 1. The cold trap couples into one of the horizontal arms of a pyrex tee fitting. The top arm of this tee fastens to the under side of the base plate. The bottom arm fastens to the top side of the mounting table. The working section of the equipment is enclosed by the main tube which is a section of pyrex pipe with an inside diameter of six inches and a length of four feet. The main tube couples into the base plate on the bottom and is capped on the top by the end plate. Attached to the end plate is the cable running to the counterweight. The counterweight facilitates easy removal of the main tube.

All of the glass components of the vacuum system are made of pyrex glass and were obtained commercially. It was decided that pyrex glass should be used because of the thermal stresses which would certainly be set up during operation. All joints in the vacuum system, with the exception of the connections beneath the mounting table, are of the simple compression type wherein two surfaces are forced together upon a soft rubber gasket. These joints were exceptionally tight and gave very little trouble.

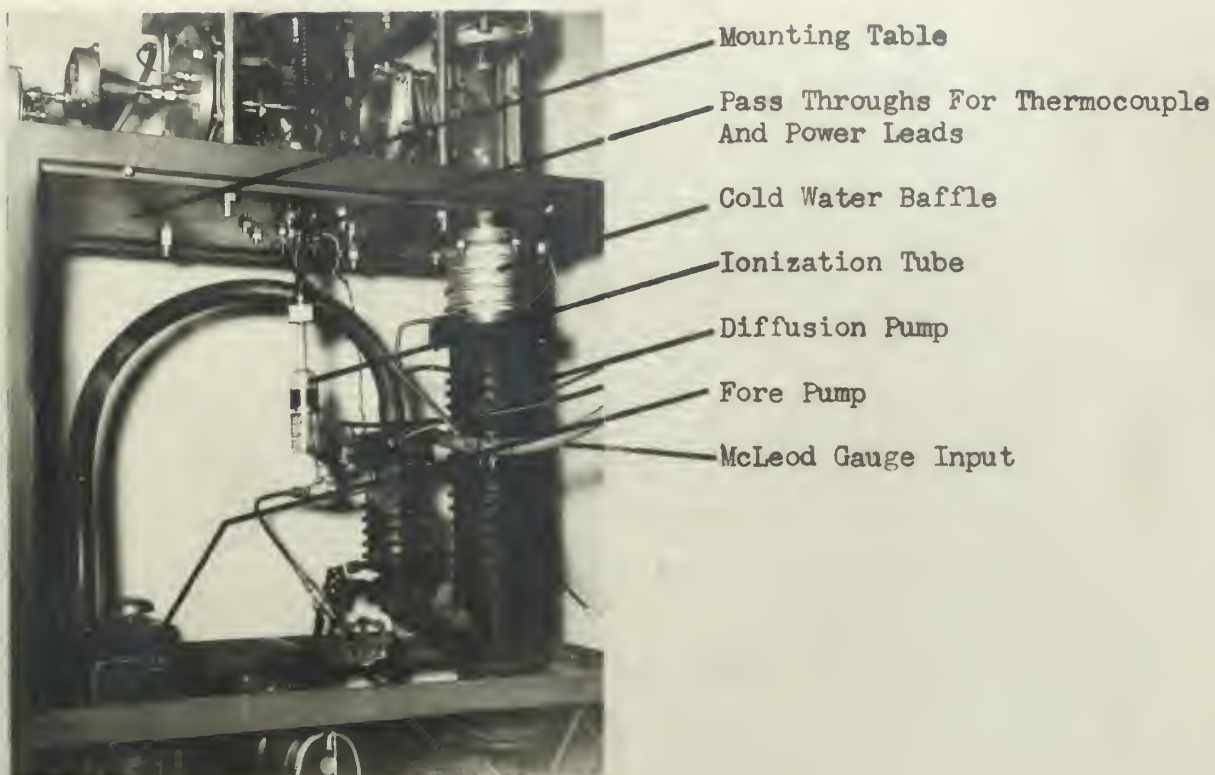


Figure 3
Lower System Components

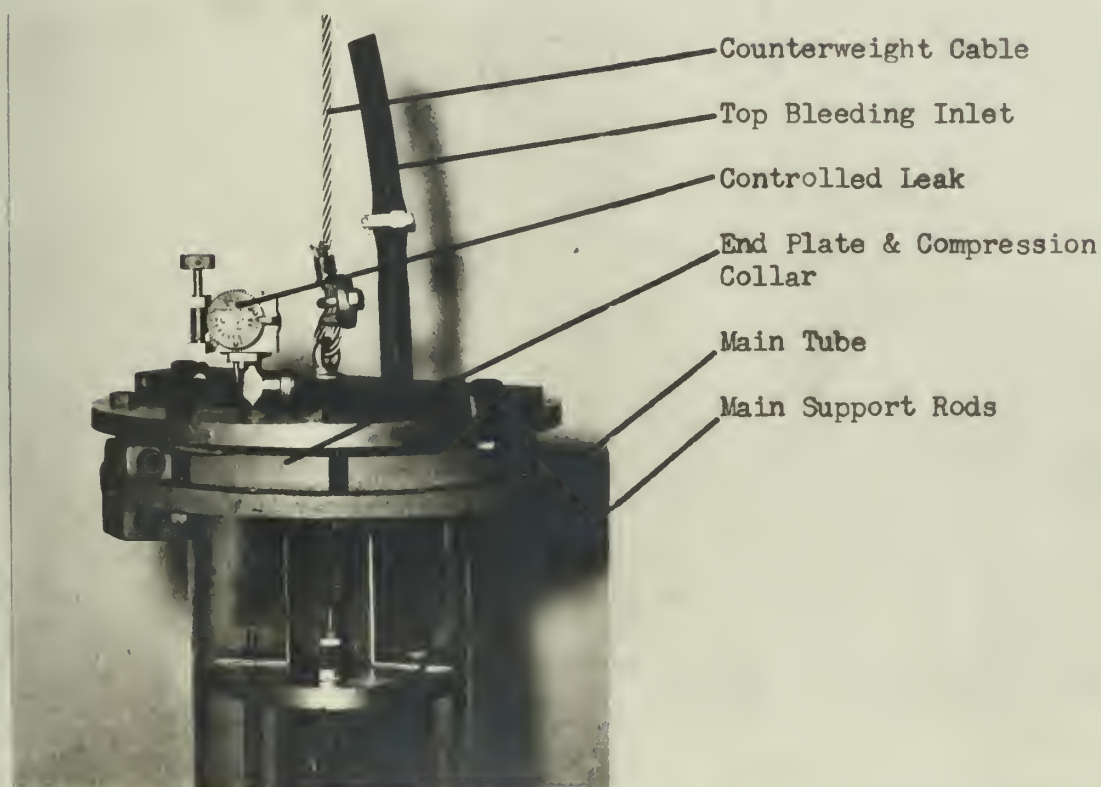


Figure 4
Top Section Of Main Tube

Provisions were made for varied methods of metering the vacuum. A manifold with two outlets is located on the high pressure side of the diffusion pump. To one of these outlets is attached the input to the McLeod gauge located on the control console. The other outlet of the manifold was coupled into a Pirani gauge while the system was being tested. The Pirani gauge was used to detect large leaks in the vacuum system and was removed when these leaks had been corrected. The primary method of vacuum determination is the ionization gauge. The ionization gauge itself is located on the control console. Its detector, the ionization tube, passes into the vacuum system through the mounting table into the lower arm of the glass tee. See Fig. 3 for these details.

Located on the end plate shown in Fig. 4 are two more items connected to the vacuum system. The first is the top bleeding inlet which is used to bleed the system after operation. It is located here to prevent the possibility of drawing oil vapor from the diffusion pump into the working section when the system is bled to atmospheric pressure. The second item is the controlled leak. Its function in the equipment is twofold. First, it provides a means of setting the pressure in the working section at any desired level. Second, after the system has been completely pumped down, one can bleed an inert gas through the controlled leak up to any desired pressure. Thus, one can obtain an inert atmosphere in the working section of known composition and pressure.

After the vacuum system had been tested and adjusted initially, it proved very reliable. The best vacuum obtained,

using liquid nitrogen in the cold trap, had a pressure of 2×10^{-5} mm. of Hg. However, this was substantially better vacuum than required, so during subsequent operation, the liquid nitrogen was omitted from the cold trap. Operation in this manner consistently gave pressures of about 3×10^{-5} mm. of Hg. It was at this pressure that most of the experimental runs were made. Normally, it takes about 45 minutes to an hour to pump the system from atmospheric pressure down to 3×10^{-5} mm. of Hg.

3. Mechanical System

As mentioned in the previous chapter, the equipment was designed with a stationary furnace and a mobile specimen. The specimen is attached to a drive chain which pulls it through the furnace. The drive section of the system is shown in Fig. 6. The drive motor operates on 110 volt, 60 cycle current, and due to a built in gear reducer, has an output shaft which turns at one revolution per second. The output of the drive motor is geared into the input of a 200 - 1 reduction gear. In turn, the reduction gear is coupled into the input of a variable reduction gear. The gear ratio of the variable reduction gear may be changed continuously from 6 - .1 to 1 - 6, and has connected in series a constant 20 - 1 gear reduction. The variable gear ratio is used to provide a continuous distribution of drive speeds. Next, the variable reduction gear is coupled to the drive shaft which passes through a rotary seal into the vacuum and terminates in a one inch diameter drive sprocket. The drive shaft and drive sprocket can be seen in the left arm of the glass tee in Fig. 1.

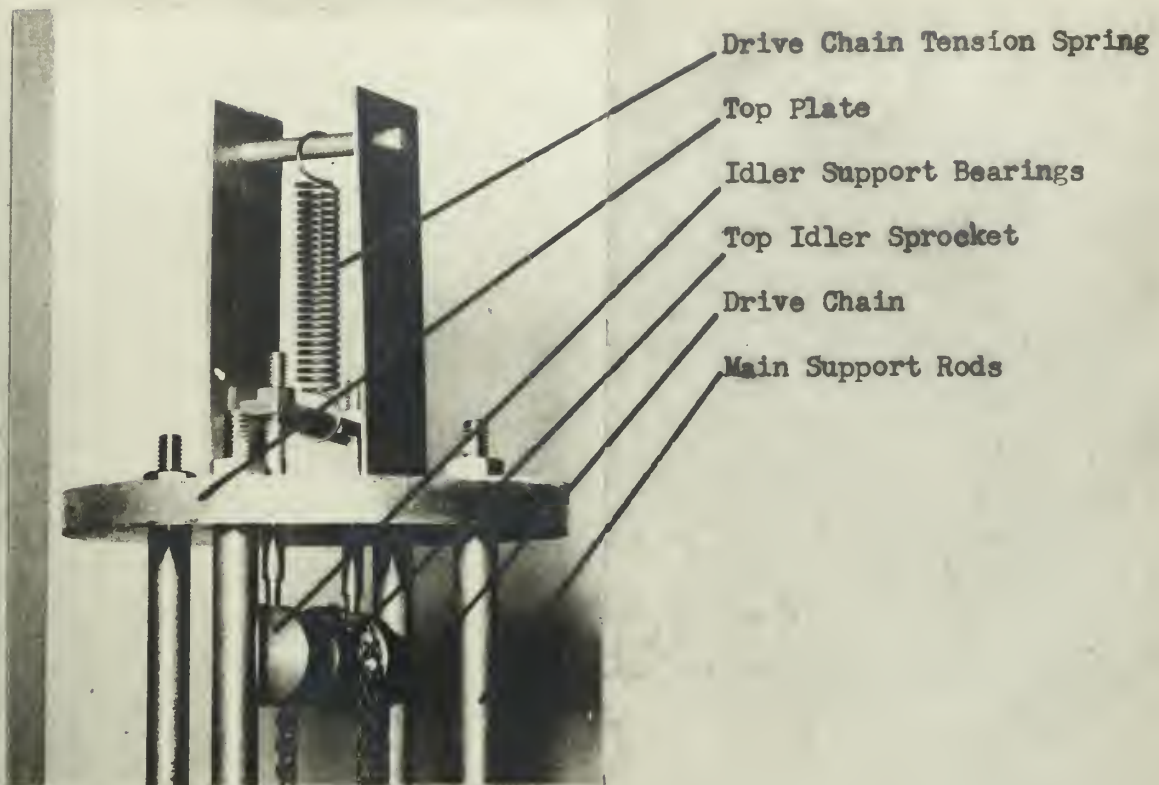


Figure 5
 Top Section Of Drive Assembly

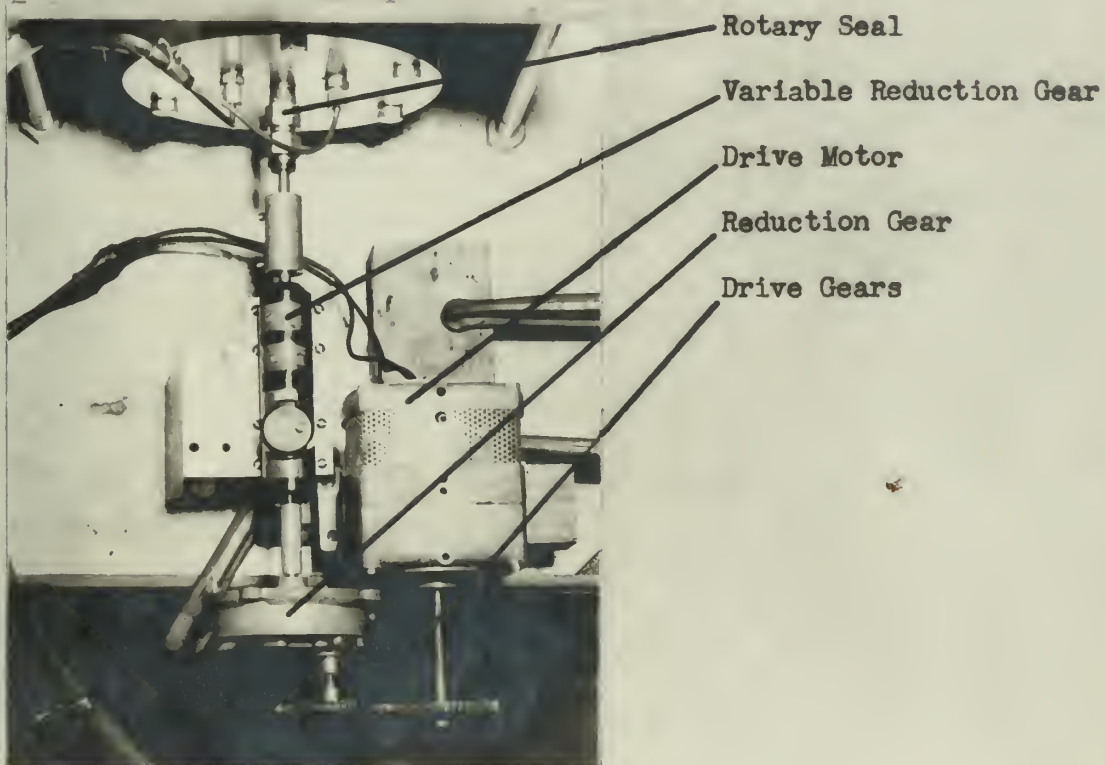


Figure 6
 Drive System

The drive chain passes around the drive sprocket, up through the working section and over the top idler sprocket. Details of the drive chain can be seen clearly in Fig. 5 and 8. The top idler sprocket is supported by a yoke which passes through the top plate. The yoke is in turn connected to the drive chain tension spring. The function of this assembly is to provide constant tension on the drive chain and to remove any slack induced by thermal expansion.

Fig. 7 and 8 shows the specimen grip. The specimen is fed through the furnace and is attached to the top specimen grip. As the drive chain moves, the specimen is pulled up through the furnace.

4. Furnace

The furnace is located in the center of the working space and is mounted on one of the main support rods. Details of furnace construction and of the surrounding items can be seen in Fig. 7 and 8. The furnace itself consists of a tungsten heating coil wrapped around a ceramic furnace insert located inside a hollow, ceramic furnace sheath. The furnace sheath has two purposes. It serves as a means of mounting the furnace insert without danger of shorting the heating coils, and also acts as a radiation shield to minimize radiation loss from the furnace. The furnace sheath passes through two steel furnace support blocks and is secured there by Allen screws through the blocks. The furnace support blocks are fixed to a rectangular steel furnace mounting plate. This is fastened to an identical plate by furnace positioning screws located at each corner. The second furnace mounting plate is attached to

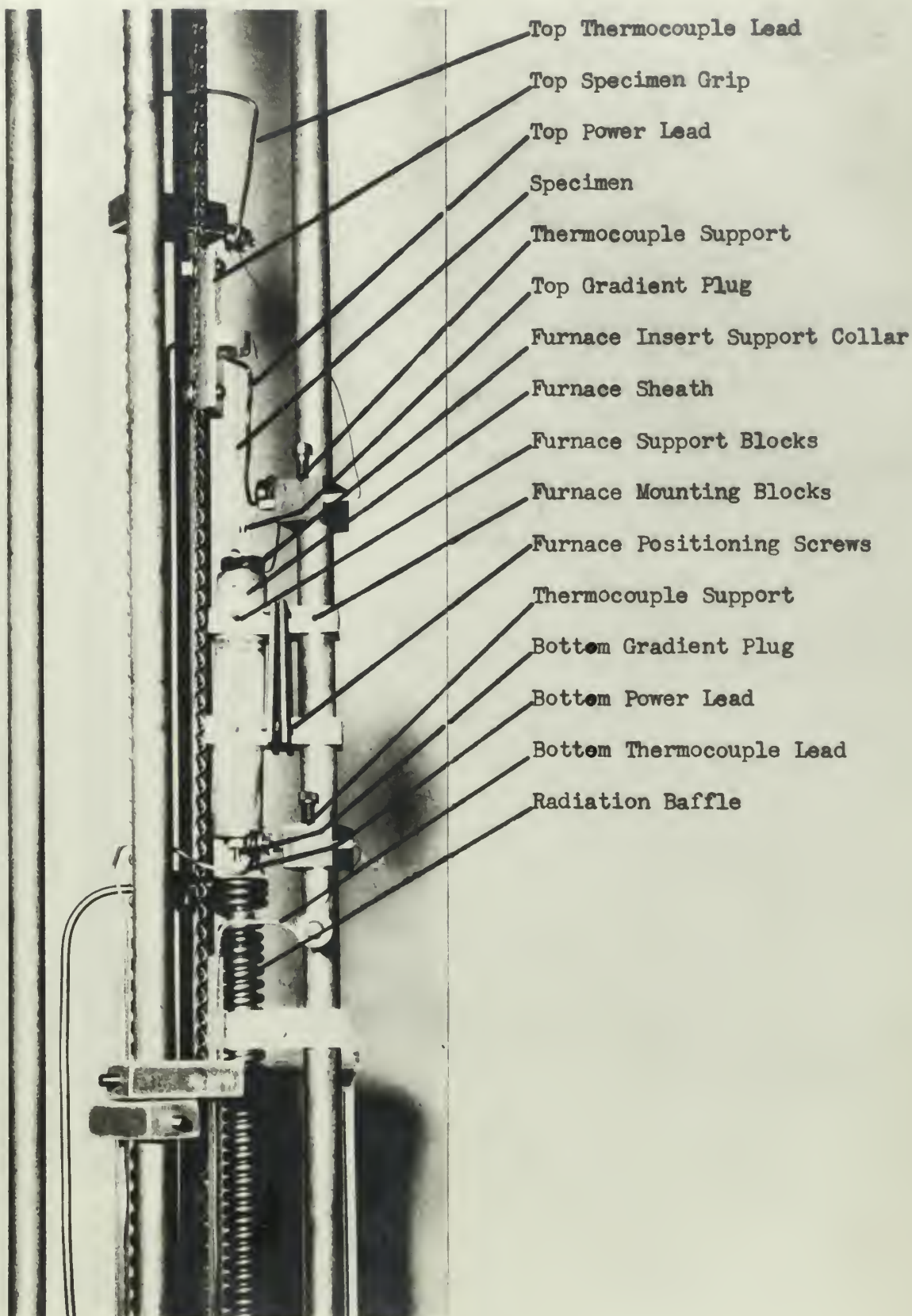


Figure 7
Furnace Section Detail

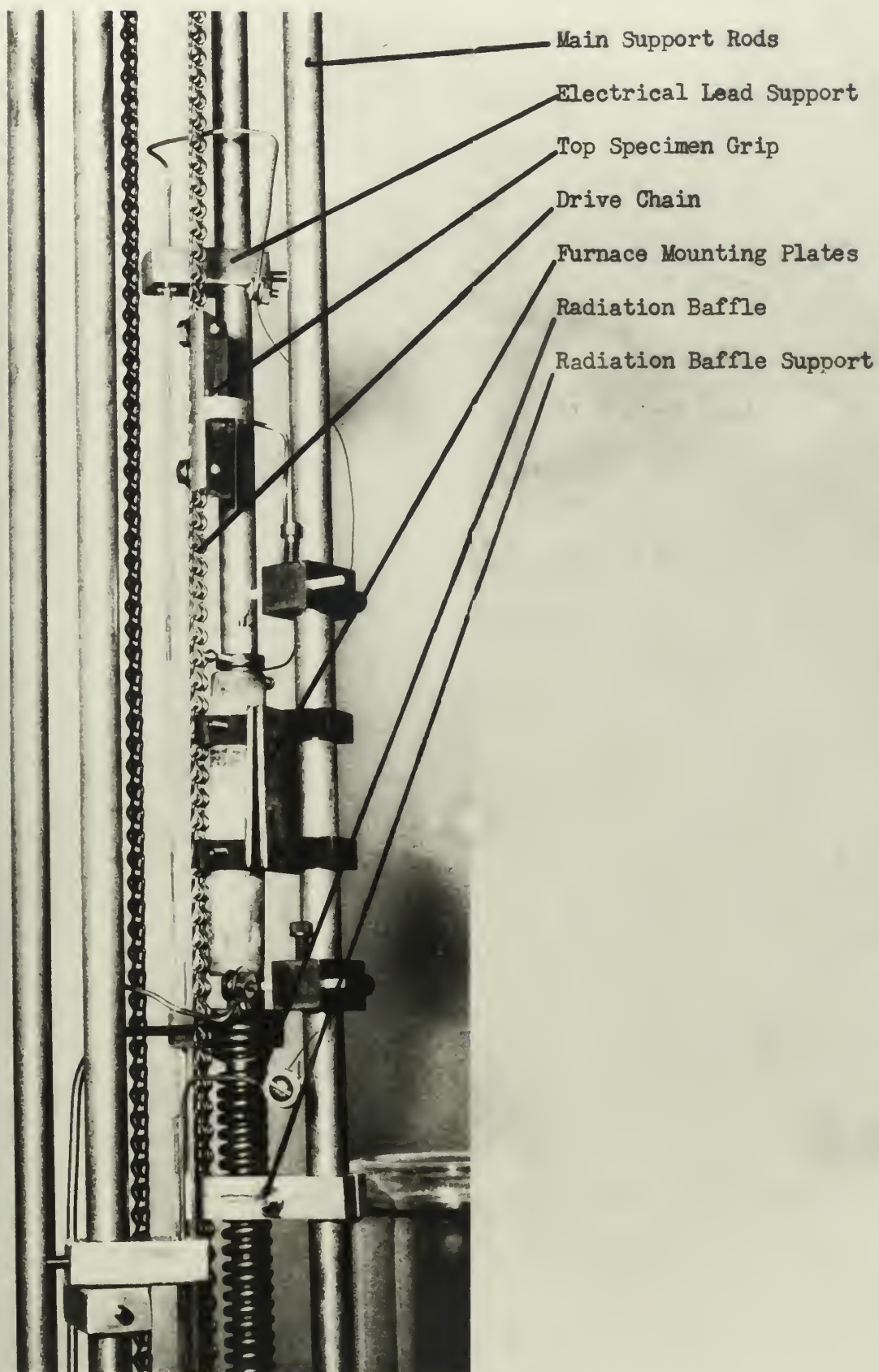


Figure 8
Furnace Section Detail

the furnace mounting blocks. One of the main support rods passes through these blocks. The furnace mounting blocks are secured to the main support by Allen screws.

Thus, the furnace can be positioned vertically by sliding the whole assembly up and down the main support rod. It has an additional degree of freedom in that the furnace may be rotated about the main support rod. Fine adjustment can be obtained with the four furnace positioning screws.

The furnace insert is a ceramic rod one-fourth inch in diameter and four inches long. It contains two holes $\frac{3}{32}$ inch in diameter which are parallel to the axis of the rod. The specimen passes through one of these holes and the thermocouple passes through the other. The furnace insert is required for two important reasons. Most important, it forces the specimen and thermocouple to maintain constant orientation with respect to each other and also with respect to the heating coils. Also, the furnace insert serves as a thermal buffer to prevent sudden changes in temperature. This last feature makes it comparatively easy to adjust the temperature and gives good thermal stability. The furnace insert is held in place by the furnace insert support collar. The insert hangs from this collar down through the furnace sheath. The tungsten heating coil is wound directly upon the furnace insert. Fig. 7 shows the ends of the heating coil emerging from both ends of the furnace sheath.

Due to the problems of outgassing, insulated wire was not used in the working section of the equipment. Instead, bare

copper wires are encased in glass tubing to insulate them from the metal portions of the system. The electrical pass-throughs which go from the under side of the mounting table into the bottom arm of the glass tee are shown in Fig. 3. The furnace and thermocouple leads go from there up through the base plate to the vicinity of the furnace. These leads can be seen in Fig. 7 and 8. Fig. 7 and 8 show the manner in which the thermocouple leads are brought through the furnace insert and thermocouple supports. The thermocouple is 13 mil chromel-alumel. It is adjusted so that it has the same geometrical relationship to the heating coils as the specimen, and hence, measures specimen temperature.

The thermocouple, using an ice-water bath as a standard reference, is balanced against a precision potentiometer for calibration. Then, the thermocouple output is fed to a self balancing recorder that is calibrated to read directly in degrees centigrade. This provides a means by which a continuous record of the specimen temperature can be maintained if required.

It was found on some of the longer runs which were made that the furnace has a remarkable degree of thermal stability considering the fact that there is no automatic temperature regulating mechanisms used in the control equipment. If care is used in adjusting the power fed to the furnace for the first two hours, thermal equilibrium is reached, and no further adjustment is required for the remainder of the run. In some cases, this was as long as 22 hours. During this time, the

temperature varied less than five degrees from the value to which it was initially set.

5. Temperature Gradient

As was mentioned in the previous chapter, one of the requirements for single crystal specimen growth is a sharp thermal gradient across the ends of the furnace. There are two devices built into the equipment that primarily serve this end. Examination of Fig. 7 and 8 will show a metallic plug extending from either end of the furnace insert. These are the top and bottom gradient plugs made of copper and tapered to fit into the axial hole through the furnace insert which contains the specimen. The gradient plugs have a 0.040 inch hole drilled through them to allow the specimen to pass through freely. The gradient plugs serve as heat reservoirs which tend to sharpen the thermal gradient across the ends of the furnace insert. Also, they force the specimen to stay along the axis of the hole through the furnace insert. This is required since the specimen cannot be permitted to touch the wall of the insert. Thermal considerations require that the temperature of the insert exceed that of the specimen. As will be seen in Chapter IV, specimen temperatures approaching the melting point of copper were required for proper crystal growth. At these temperatures, contact between the specimen and the insert would result in immediate melting and destruction of the specimen. Also, contact may allow impurities to transfer from the ceramic insert to the specimen. However, both these possibilities are eliminated with the gradient plugs.

The second device to produce a sharper thermal gradient is the radiation baffle shown in Fig. 7 and 8. It consists of a section of large copper tubing upon which has been wrapped a cooling coil of one-eighth inch copper tubing. On the end of the radiation baffle, there can be seen a section of extended coils. These pass around a large copper plug which was machined to fit in the end of the copper tubing which forms the core of the radiation baffle. This plug has an axial 0.040 inch hole drilled through to allow the specimen to pass through freely. The ends of the cooling coil are brought out of the vacuum along the drive shaft and can be seen in Fig. 5. In most of the runs which were made, the coolant was cold water. The radiation baffle is mounted immediately below the furnace and is adjusted so that the specimen can be threaded through the furnace and down into the baffle. A set of weights were made which can be attached to the specimen inside the radiation baffle. This permits varying amounts of tension to be put on the specimen.

The radiation baffle serves various roles depending upon the manner in which the runs are made. Usually the specimen is adjusted so that it just touches the plug in the top of the radiation baffle. Since this plug is kept at a low temperature, any heat which travels down the specimen from the furnace is drawn off here. The rest of the baffle forms a barrier against thermal radiation. If the run is made in hard vacuum, this is its only function. However, if the run is made in an inert atmosphere, the radiation baffle can exert a more active role

in maintaining low specimen temperature on that portion which has not passed into the furnace. This of course, is due to the convection currents which are set up in the interior of the baffle.

Examination of the experimental results indicates that the combination of the gradient plugs and the radiation baffle gave a fairly sharp thermal gradient as was desired.

6. Control Console and Safety Considerations

Fig. 9 shows a front view of the control console and the temperature recorder. On it can be seen the various portions of the control equipment. Fig. 10 shows the control system schematic. Two high power variacs are used to drive the furnace. The control system can put a maximum of 15 amperes through the furnace heating coils. It was found out that this current resulted in a specimen temperature of over 1200 degrees centigrade. Since this is well over the melting point of copper, the temperature capabilities of the furnace are sufficient. The auxiliary power outlet is used to power the fore-pump. Therefore, all components of the control system with the exception of the ionization gauge, feed through the main power switch.

Located in the ionization gauge is a relay set to operate if the pressure in the system rises above 10^{-3} mm. of Hg. When this happens, it activates the relay shown in Fig. 10. This action cuts off the diffusion pump before destructive distillation of the diffusion pump oil occurs.

The diffusion pump has a feature which makes for safe

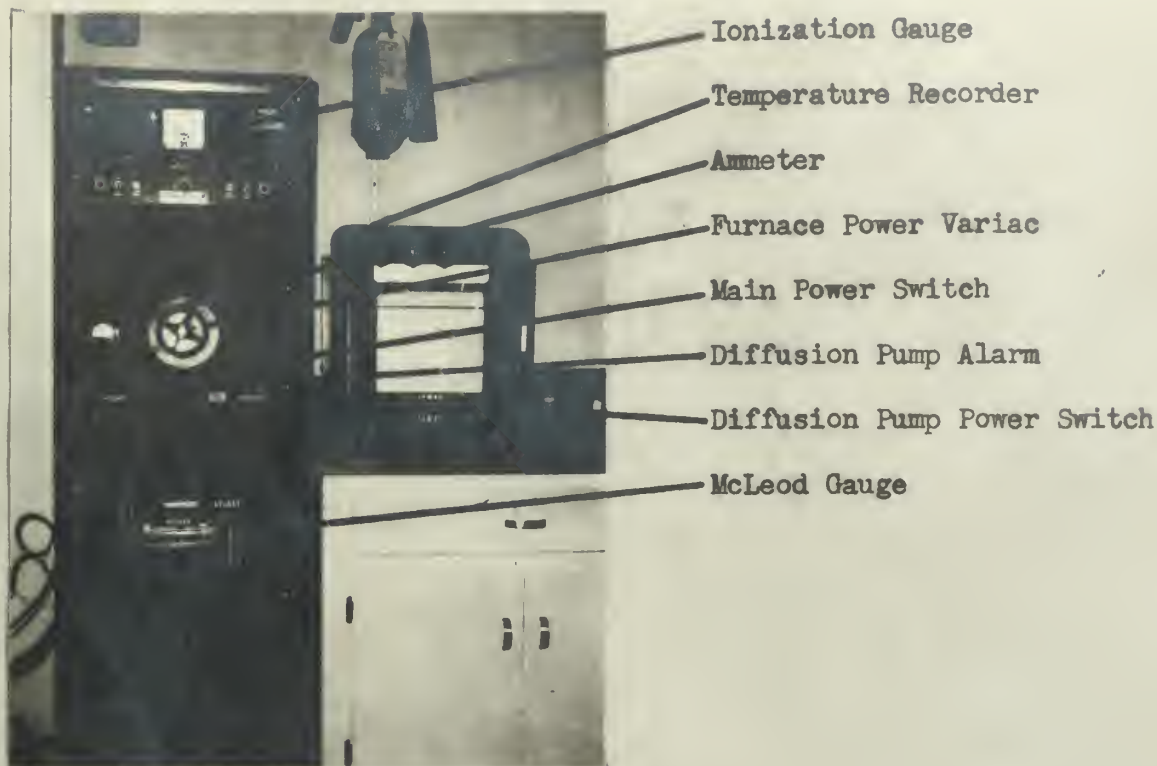


Figure 9
Control Console

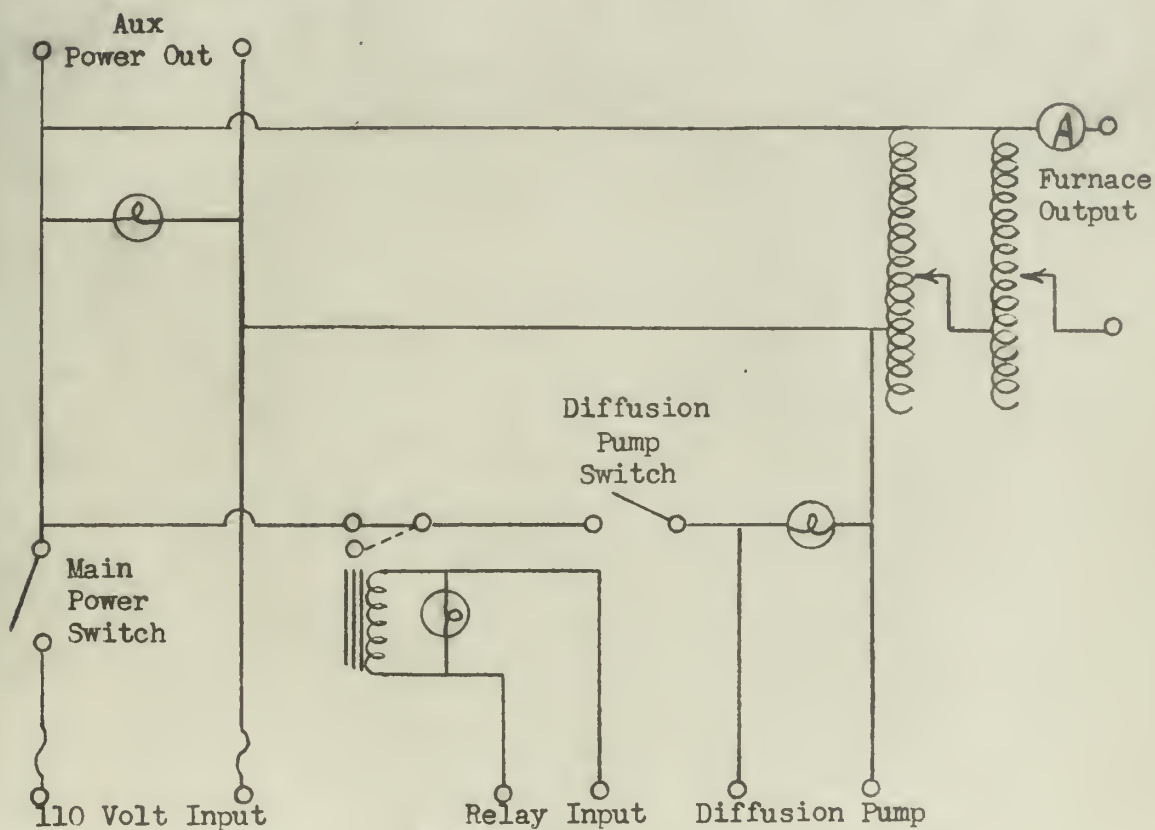


Figure 10
Control System Schematic

operation. A valve system permits cold water to be sent through either the normal cooling coils of the diffusion pump, or through the quenching coils located on the bottom. After a run, cold water is forced through the quenching coils to cool the oil inside the diffusion pump rapidly. This allows one to open the system immediately without the danger of ruining the diffusion pump oil by pyrolysis.

When longer runs were made on the equipment, it was noticed that the portion of the main tube in the vicinity of the furnace got very hot. It was felt that there was a possibility that either through thermal causes, or through a mechanical shock which could be delivered accidentally, the main tube might implode. Therefore, an implosion shield was installed to protect personnel in the event of such an occurrence. Fig. 11 shows the equipment with the implosion shield in place.

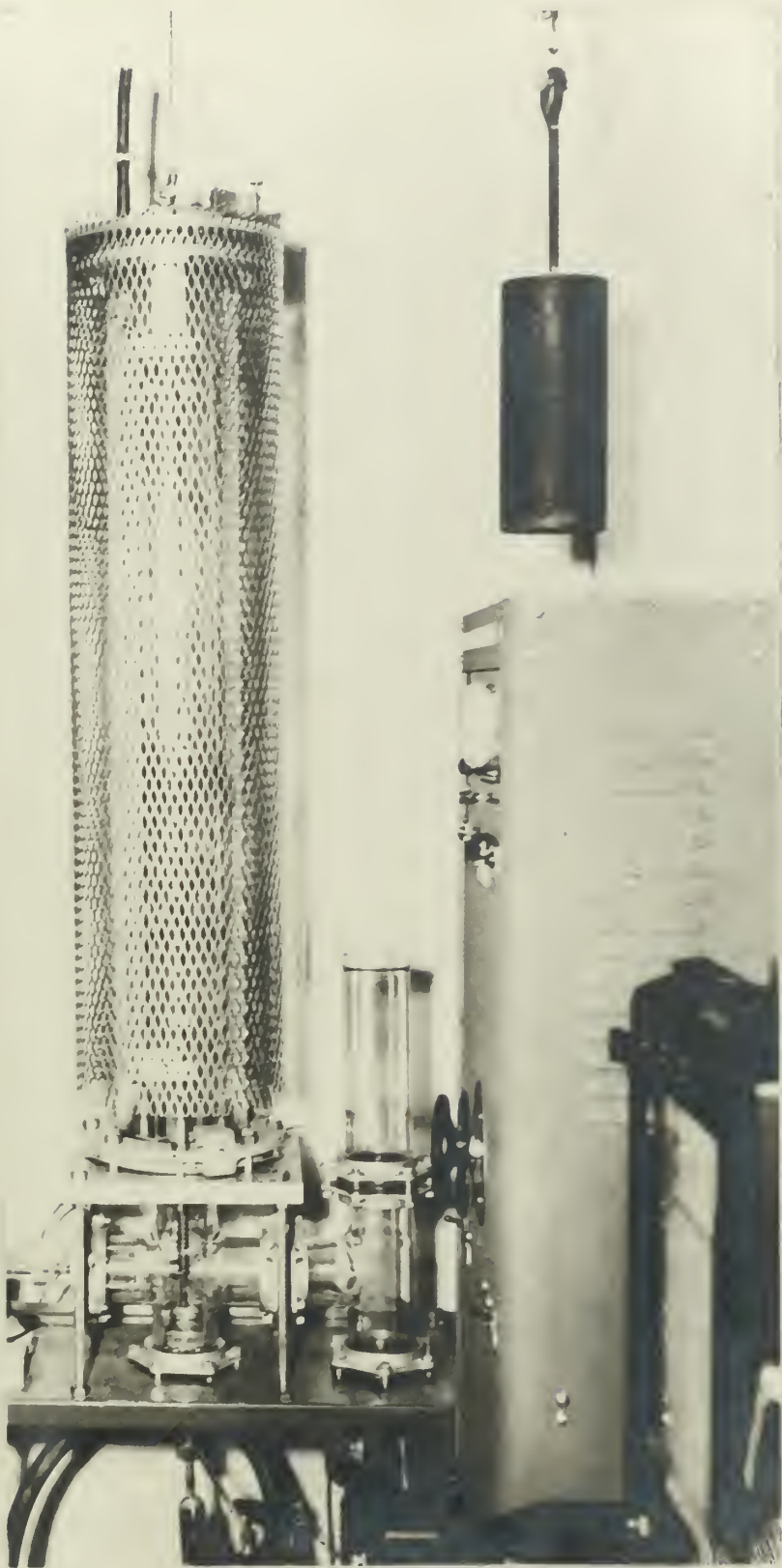


Figure 11
Equipment With Safety Shield In Place

CHAPTER III

TESTING OF EQUIPMENT

Preliminary design of the apparatus was completed in June 1956 and sundry portions of the equipment were fabricated by the metal shops of the Postgraduate School during the months of June and July 1956. Construction proceeded during the period September - November 1956 and the equipment was ready for its first vacuum test in December. The usual vacuum difficulties were encountered requiring the replacement of a poorly welded diffusion pump, and several complete disassemblies of the entire system. A working vacuum of 3×10^{-5} mm. of Hg., was achieved during the first week of January 1957, and the furnace construction began.

Proof testing of the system as a whole commenced on 15 January 1957 with the first of 28 runs used to complete preliminary testing. This run aborted when the thermocouple shorted out several coils of the tungsten heating element and destroyed itself. At this stage of development, the furnace did not contain an insert and there was no method of preventing the thermocouple or the specimen from bowing or swinging into contact with the heating element.

A new furnace was constructed with the furnace insert described in Chapter II. In addition, specimen gradient plugs were added to intensify the natural gradient at the ends of the furnace insert.

A new chromel - alumel thermocouple was constructed with a butt-weld junction approximately 1/16 inches in diameter to facilitate mounting in the furnace insert.

Both a top and a bottom specimen grip were used on these early runs. Friction between the specimen and the bottom grip negated the effect of the tension weight hanging on the specimen. This was evidenced by bowing between the bottom specimen grip and the lower end of the furnace. After the first six runs, the bottom specimen grip was removed and the specimen allowed to hang free.

Completion of proof testing required 23 additional runs. Speeds ranged from eighteen to three centimeters per minute, and indicated temperatures ranged from 800 to 1240° centigrade. Temperature as recorded was highly erratic as successful runs were made at an indicated 1240° centigrade, some 157 degrees above the melting point of copper, while other specimens melted at an indicated 970 degrees, or 113 degrees below the melting point. It became obvious that finer thermocouple adjustment was needed to correct this situation, so top and bottom thermocouple gradient plugs were added and tension was maintained through the thermocouple supports. This tension insured that the thermocouple did not touch the walls of the insert and resulted in accurate and reproducible temperature indications.

The radiation baffle was added to give a sharper thermal gradient at the lower end of the furnace and was successfully used with both cold water and helium precooled in a liquid nitrogen bath as coolants.

At this stage, the apparatus seemed to be working properly and giving reproducible speeds and temperatures with satisfactory stability on long runs, so the series of experimental runs described in Chapter IV commenced on 19 February 1957.

CHAPTER IV

EXPERIMENTAL PROCEDURES

The experimental procedures envisaged for the investigation of single crystal growth in the vacuum annealing furnace consisted of a systematic evaluation of the variables involved. It was planned to make the first few runs with constant vacuum, speed, and thermal gradient. Temperature and tension would be varied. A thorough examination of the results of these runs should reveal some optimum combination of temperature and tension which would be used in the second series of runs in which specimen velocity would be varied. The combined results of the first two series of runs should indicate the next variable to be investigated.

For an experimental run, loading was accomplished as follows. A length of the cold-drawn copper wire was selected and cut to 60 cm. The tension weight was fastened to one end, and the radiation baffle plug and bottom gradient plug were threaded on. The main tube was raised, the radiation baffle supports loosened, and the tension weight and plug were inserted in the radiation baffle. By means of a wire hook, the specimen was then pulled through its channel in the furnace insert and the top gradient plug was threaded. The top of the specimen was then fastened to the top specimen grip. The radiation baffle was aligned so that the specimen contacted the plug in the top of the radiation baffle to insure thermal conduction. Then

the radiation baffle supports were tightened. The top and bottom gradient plugs were wedged into the furnace insert and the thermocouple supports adjusted for tension. The drive chain tension device was adjusted, and then as a final step, the main tube was lowered and secured in place. The apparatus was then ready for evacuation.

The first series of experimental runs were made at a pressure of 3×10^{-5} mm. of Hg. and at a specimen speed of 0.54 millimeters per second. Approximately eight centimeters of specimen were annealed at 900° centigrade, then a few centimeters were pulled through as the temperature was raised to 950° C. Eight centimeters of specimen were annealed at this temperature, then eight more at 1000° C. and finally, eight centimeters at 1020° C. This last temperature was found to be the maximum attainable without frequent abortions caused by specimens melting during the run.

Three runs, which are described in detail above, were carried out using 23 grams tension, 11 grams tension, and no tension on the specimen. Samples of wire approximately one-half inch long were taken from that portion of each temperature section which was within the radiation baffle at the start of its temperature and above the furnace before the temperature was raised. These were mounted longitudinally on the surface of one inch diameter, one-half inch thick bakelite plugs in a thermosetting compression press. The sample was ground down until it had been halved longitudinally and then polished on a one-micron diamond polishing wheel. The surface was etched

with a solution of two grams of $K_2Cr_2O_7$ and eight milliliters of 1.84 specific gravity H_2SO_4 in 100 milliliters of water. Four drops of concentrated HCl were added to this solution immediately before etching. An etching time of approximately ten seconds was used. Microscopic examination of the sample was used to select a section exhibiting the typical grain size resulting from the temperature-time combination used. This section was microphotographed and the pictures used for comparison of the various runs.

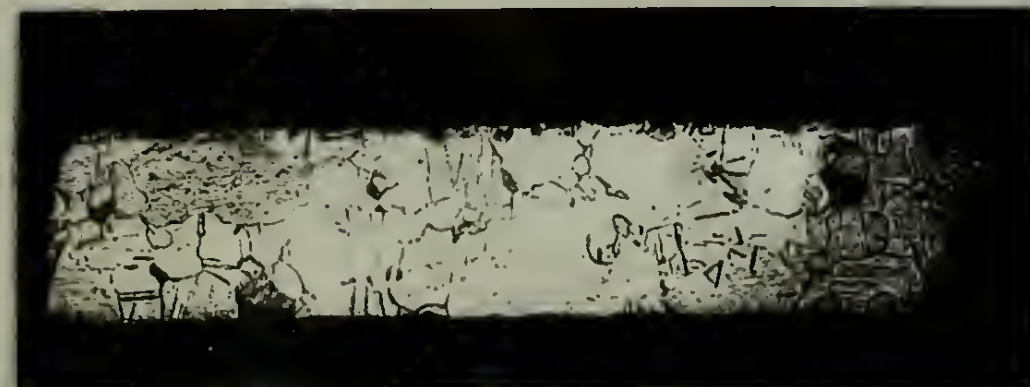
Additional samples were examined by x-ray diffraction techniques and the expected preferential $[111]$ orientation was confirmed.

The microphotographs picturing the results of the first experimental series appear on Fig. 12. A detailed examination of this figure reveals that the average longitudinal crystal dimension increases steadily with increasing temperature and with decreasing tension. The best specimen produced in this series, Fig. 12(d), represented zero grams tension and 1020° centigrade.

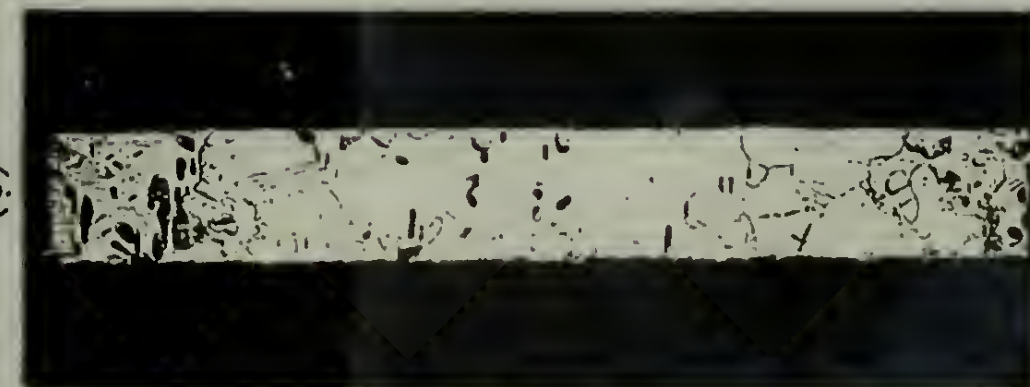
The next variable investigated was specimen velocity through the furnace. Preliminary runs revealed that a minimum tension of 2.5 grams was necessary at very slow velocities to prevent bowing or thermal sagging with subsequent melting of the specimen, so this tension was used throughout. Also, approximately 50% of the runs with this tension melted at 1020° C., so a temperature of 1010° C. was decided upon.



0 Grams Tension



11 Grams Tension



23 Grams Tension



(b)



(f)



(j)
950° C.



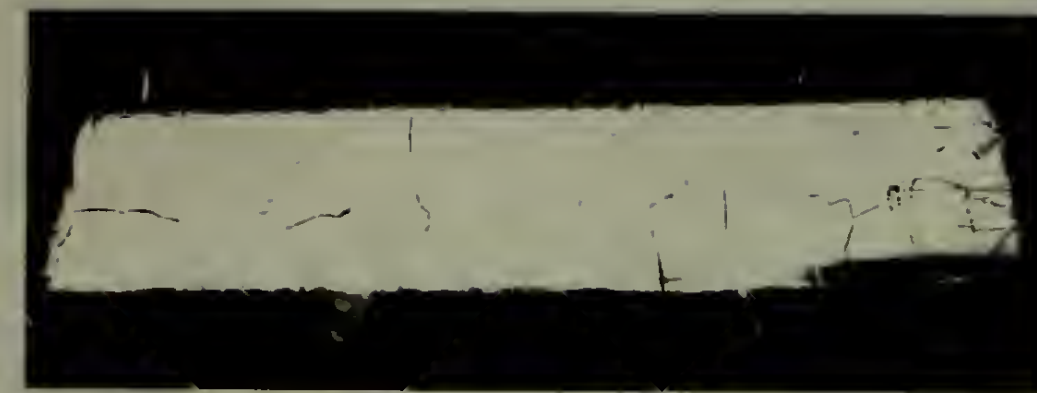
(c)



(g)



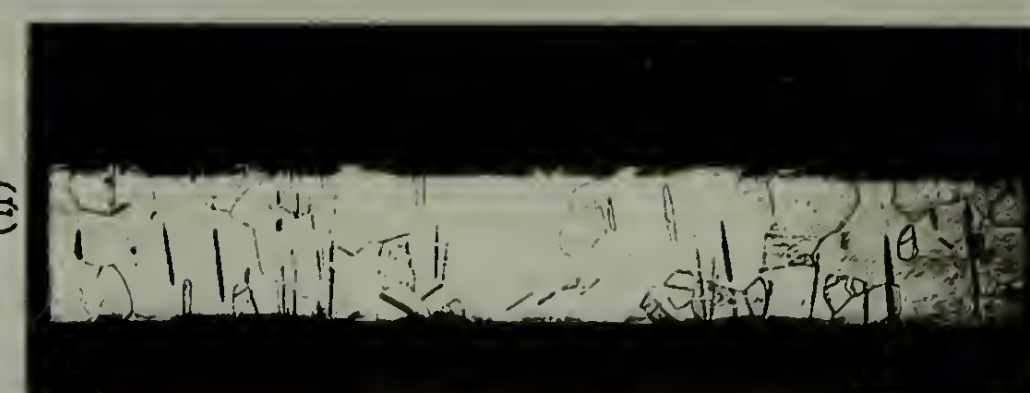
(k)
1000° C.



(d)



(h)



(l)
1020° C.

Figure 12
Specimen Microphotographs

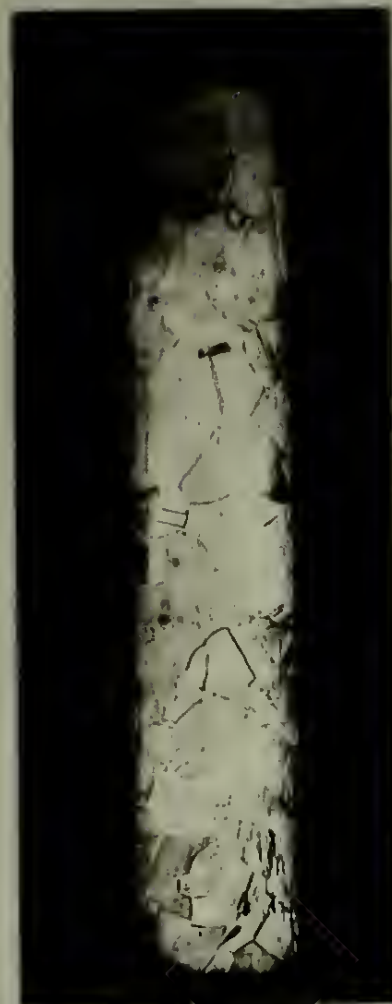
Twelve different velocities were investigated at 1010° C. and 2.5 grams tension. Microphotographs were taken and appear in Fig. 13. The speeds represented in this figure are:

(a) 2.7 mm./second	(g) 0.18 mm./second
(b) 1.8 "	(h) 0.15 "
(c) 0.90 "	(i) 0.03 "
(d) 0.45 "	(j) 0.015 "
(e) 0.30 "	(k) 0.008 "
(f) 0.23 "	(l) 0.005 "

The average crystal length along the fiber is seen to increase steadily with decreasing speed. The best sample, Fig. 13(1), represents specimen velocity of 1.8 centimeters per hour through the furnace. This velocity is the lower limit for the gearing system used. Since about 75% of the runs made at this speed and temperature aborted after ten to thirty centimeters of specimen had been annealed, it appears that some means of automatic temperature control may be a requirement for still slower specimen speeds with this apparatus.



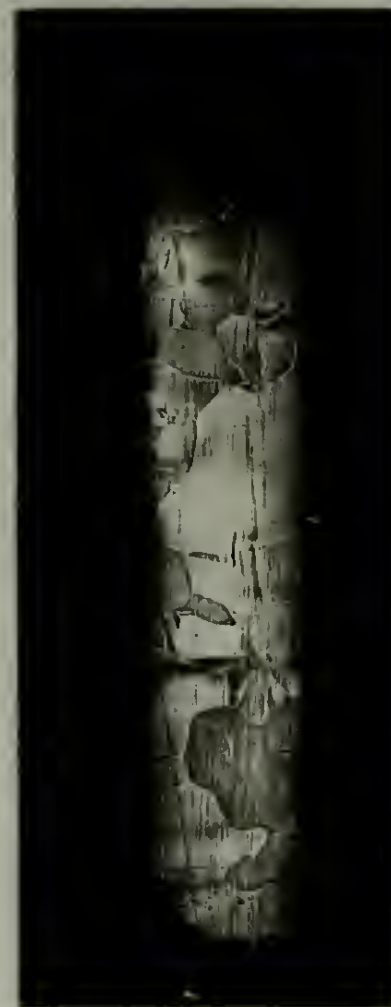
(a)



(b)



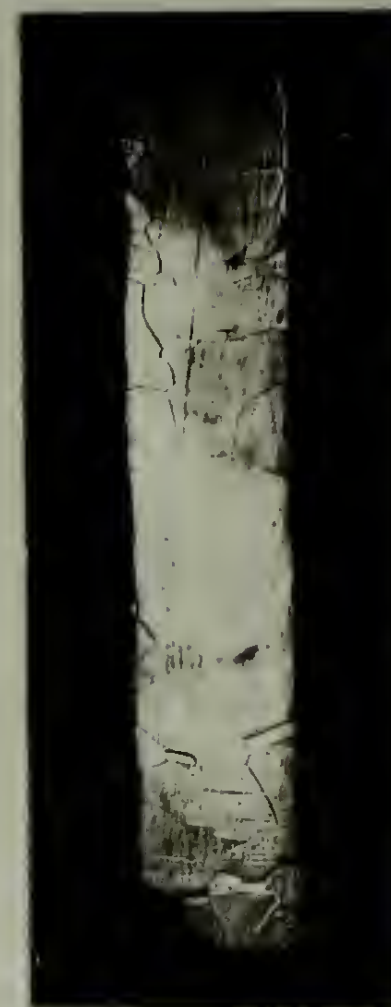
(c)



(d)



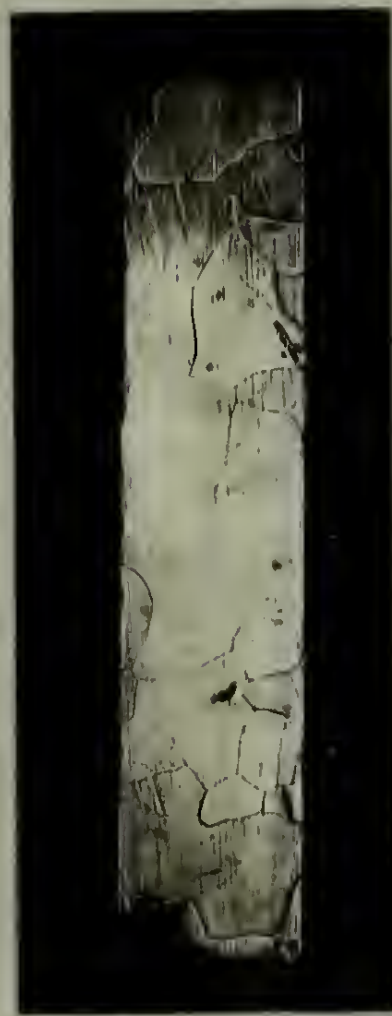
(e)



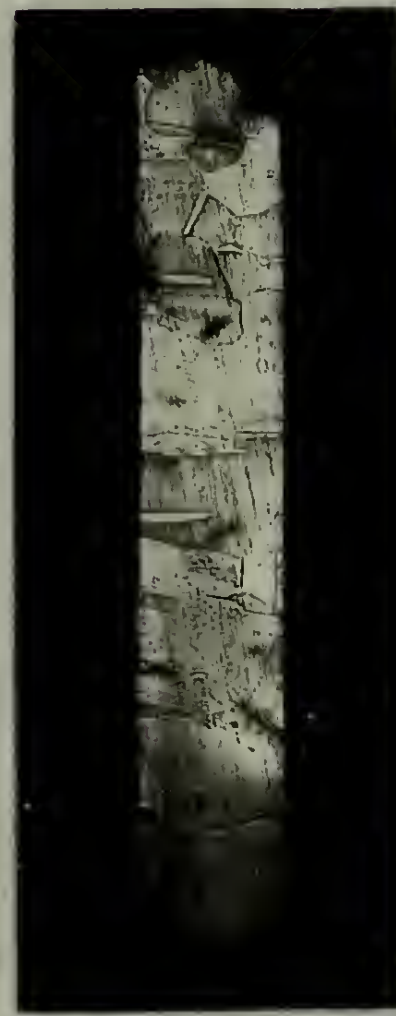
(f)



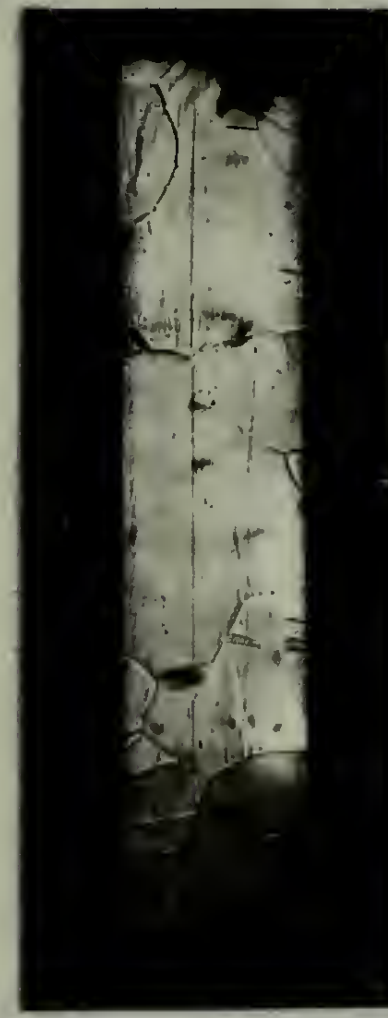
(g)



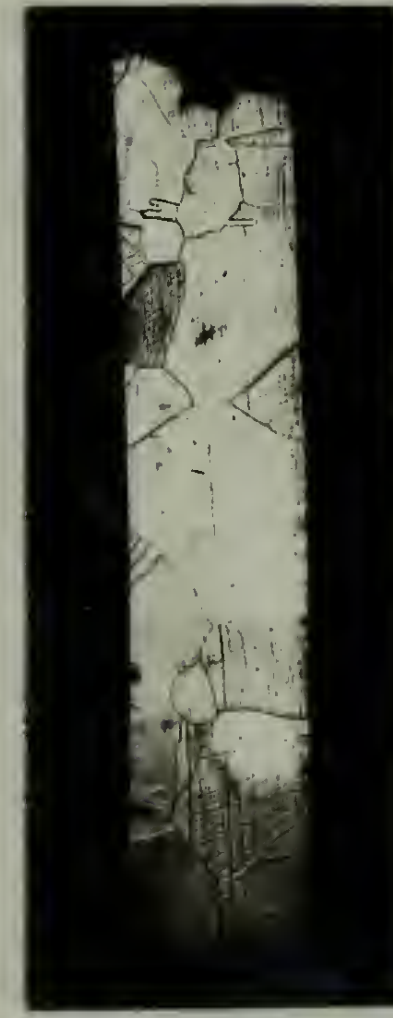
(h)



(i)



(j)



(k)



(l)

Figure 13
Specimen Microphotographs

CHAPTER V

DISCUSSION AND CONCLUSIONS

The specimens produced under the conditions at which the specimen pictured in Fig. 13 (1) was made, show a considerable improvement over the ones which were used previously in the investigation of irradiation effects upon the shear modulus of copper. Thus, it is felt that the apparatus as constructed is sufficient to produce adequate specimens to further investigations in this field. However, there are still significant numbers of crystal imperfections and boundaries in the best specimens which were produced. It appears, however, that if time were available to complete the systematic investigation of the variables in the problem and to complete further refinements of the apparatus, single crystal copper fibers of any desired length could be produced.

Several runs were made under the conditions with which the best specimen was produced. Results of these runs essentially duplicated the longitudinal crystal lengths previously obtained and confirmed that specimens of this degree of crystal purity can be obtained routinely.

It is to be noted that three of the variables in the problem were held constant throughout the experimental procedures described. A thorough investigation of these variables should be a first step in any future attempt to refine this method of specimen production.

The first of these variables is the composition of the gaseous environment in the working space. In all experimental procedures, air was originally present and the space was then evacuated to a pressure of approximately 3×10^{-5} mm. of Hg. This was done to reduce oxidation of the specimen to a minimum. This reduction of oxidation could also be accomplished if the annealing procedure were carried out in an atmosphere of hydrogen, helium, argon or another inert gas in the working space. Provision has been made for this procedure since a controlled leak for the introduction of an inert gas was installed at the top of the main tube. The use of an inert atmosphere also allows wide variation to be made in the remaining two variables.

The second variable, the sharpness of the thermal gradient, is inextricably tied up with the composition of the atmosphere existing in the working space. The hard vacuum used in these experiments effectively prevented convection as a means of heat transfer from the specimen and greatly reduced the potential value of the radiation baffle. An inert gas at or near atmospheric pressure would allow convection in the radiation baffle to intensify the thermal gradient. The gradient may also be varied by moving the radiation baffle closer to the end of the furnace or by varying the coolant used in the cooling coils.

The third variable is the pressure used during the annealing process. Apparatus proof tests, using liquid nitrogen in the cold trap demonstrated that pressures of the order of

2×10^{-6} mm of Hg. are possible without further modification of the equipment. Use of an inert atmosphere as described in the preceding paragraph would permit runs to be made at higher pressures, even above atmospheric if desired, and should allow this variable to be thoroughly investigated.

One other major change to the equipment may well be worthy of study. This is the advisability of adding an automatic temperature control device. The simplest type of control is merely an "on-off" switch for the furnace controlled by a pre-set temperature range. This type of control has the disadvantage that the actual temperature continually "seeks" the desired temperature and many rapid fluctuations occur. There is some question about the advisability of using this type of control in the production of single crystals as the effect of these fluctuations on crystal growth is not understood. A better control would be one which varied the current through the furnace continuously. This method of control would allow for a smoother variation of temperature with time. At any rate, some method of reducing the temperature drift observed on the longer runs should be provided. It is possible that a regulated power supply for the furnace, to avoid the effects of varying line voltage during the period of operation, would be all that is required.

The investigations described herein have resulted in the following criteria which seem to maximize mean crystal length in copper fibers:

- a. Specimen temperature should be brought close to the melting point, within 60° centigrade if possible,
- b. There should be a minimum of tension used during the annealing process,
- c. The specimen velocity through the furnace should be less than 0.005 millimeters per second.

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